

REPORT DOCUMENTATION PAGE					<i>Form Approved</i> OMB No. 0704-0188	
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1. REPORT DATE (DD-MM-YYYY) 11-08-2015		2. REPORT TYPE Final			3. DATES COVERED (From - To) 16 Dec 2013 to 25 Dec 2014	
4. TITLE AND SUBTITLE Research on Electrically driven single photon emitter by diamond for quantum cryptography				5a. CONTRACT NUMBER FA2386-14-1-4037		
				5b. GRANT NUMBER Grant 14IOA093_144037		
				5c. PROGRAM ELEMENT NUMBER 61102F		
6. AUTHOR(S) Prof. Norikazu Mizuochi				5d. PROJECT NUMBER		
				5e. TASK NUMBER		
				5f. WORK UNIT NUMBER		
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) Osaka University 1-3, Machikane-yama, Toyonaka-city Osaka 560-8531 Japan					8. PERFORMING ORGANIZATION REPORT NUMBER N/A	
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) AOARD UNIT 45002 APO AP 96338-5002					10. SPONSOR/MONITOR'S ACRONYM(S) AFRL/AFOSR/IOA(AOARD)	
					11. SPONSOR/MONITOR'S REPORT NUMBER(S) 14IOA093_144037	
12. DISTRIBUTION/AVAILABILITY STATEMENT Distribution Code A: Approved for public release, distribution is unlimited.						
13. SUPPLEMENTARY NOTES						
14. ABSTRACT Nitrogen-vacancy (NV) centers in diamond have emerged as a highly competitive platform for applications in quantum cryptography, quantum computing, spintronics, and sensing or metrology.						
15. SUBJECT TERMS Diamond LED, Nitrogen Vacancy Complex, Quantum Computing, Quantum Cryptography, Single Spin Single Photon						
16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON	
a. REPORT	b. ABSTRACT	c. THIS PAGE			Seng Hong, Ph.D.	
U	U	U	SAR	11	19b. TELEPHONE NUMBER (Include area code) +81- 42*511-2000	

**"Research on electrically driven single photon emitter
by diamond for quantum cryptography"**

Date March 24th, 2015

Name of Principal Investigators (PI and Co-PIs): Norikazu MIZUOCHI

- e-mail address : mizuochi@mp.es.osaka-u.ac.jp
- Institution : Graduate School of Engineering Science
- Mailing Address : mizuochi@mp.es.osaka-u.ac.jp
- Phone : +81-6-6850-6426

Period of Performance: Dec/26/2013 – Dec/25/2014

Abstract:

Nitrogen-vacancy (NV) centers in diamond have emerged as a highly competitive platform for applications in quantum cryptography, quantum computing, spintronics, and sensing or metrology. These atomic-scale defects have two different charge states: the neutral NV^0 state, and the negatively charged NV^- . These two states have different optical and spin properties that can be exploited for applications. Deterministic and fast switching between these two states would be highly desirable. So far, however, charge-state control of the single NV center among the two charge states has only been achieved by optical means with stochastic process. Here, we demonstrate, deterministic, purely electrical, and room-temperature charge-state control of single NV^- centers. The results are useful not only for ultrafast electrical control of qubits, long T_2 quantum memory, and quantum sensors associated with single NV centers but also for classical memory devices based on single atomic storage bits working under ambient conditions. (Phys. Rev. X, 4, 01107 (2014).)

Previously, we realized electrically driven single photon source by the NV center at room temperature. However, the charge state of the NV center was a neutral charge state. The realization of electroluminescence of NV^- centers is very important because nobody has yet succeeded the electrical control of single spin at room temperature, however, it is well known that charge state of NV^- centers change to NV^0 during laser illumination. We demonstrated that doping of phosphorus generates about 99.4 ± 0.1 % NV^- . This is very important step toward electrical control of single spin at room temperature which is required for quantum repeater for long distance quantum cryptography. (Submitted to Phys. Rev. X).

Introduction:

Quantum cryptography is expected to establish a new paradigm. It is considered to be an ultimate cryptography which can not be decoded. Recently, some venture companies and big companies in Japan invest in product development research for quantum cryptography. So far, the issues are the speed and distance of communication. For faster and longer communication, devices of high quality single photon source and quantum repeater are required.

Due to the lack of a good single photon source, the present quantum cryptography systems adopt a pseudo-single photon source where the very weak light of a conventional laser is used. As for the quantum repeater, it has not yet realized even in laboratory. Long quantum memory time and processing of quantum information are required for quantum repeater, but they are not easy to realize. Therefore, basic research is very necessary for single photon emission and control of quantum information.

Recently, we investigate a NV center in diamond (Fig. 1a), which can be considered as a good candidate of the resource of them. It is because that electrically

driven single photon source at room temperature (Fig. 1c) is realized and that the single spins can be coherently controlled with long quantum memory time even at room temperature in the NV center. In other solid materials such as quantum dot or superconducting devices, they can be operated only in extremely low temperature by using liquid Helium. The unique potential of room temperature operation of the NV center is very important for the quantum cryptography.

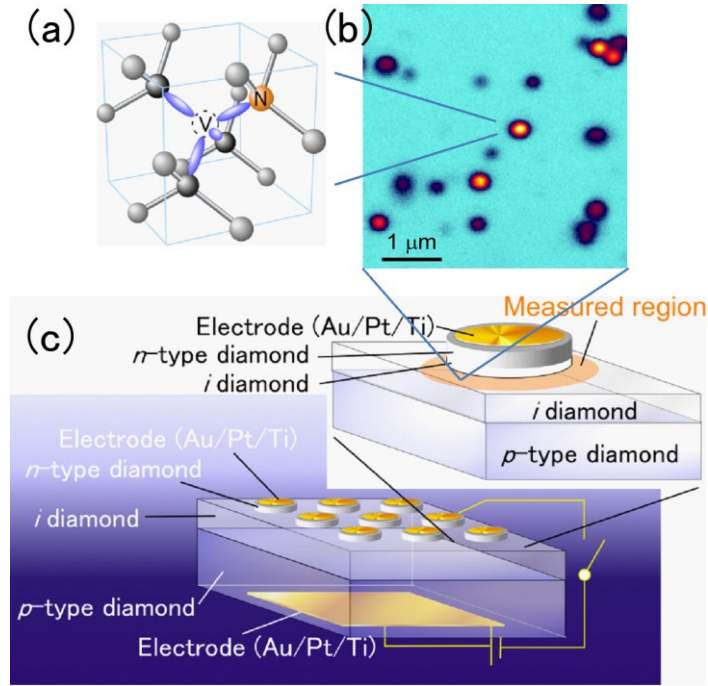


Fig. 1 (a) Structure of NV center in diamond (b) Confocal microscope image of single NV center. (c) Structure of p-i-n LED (Light Emitting Diode) diamond device.

Experiment, Results and Discussion:

(1) Deterministic electrical charge state initialization of single nitrogen-vacancy center in diamond (Phys. Rev. X, 4, 01107 (2014))

Apart from applications in classical information processing devices, the electrical control of atomic defects in solids at room temperature will have tremendous impact on quantum devices that are based on such defects. In this study, we demonstrate the electrical manipulation of individual prominent representatives of such atomic solid-state defects, namely, the negative charge state of single nitrogen-vacancy defect centers (NV^-) in diamond. We experimentally demonstrate for the first time deterministic, purely electrical charge state initialization of individual NV centers (Fig. 2). The NV centers are placed in the intrinsic region of a *p-i-n* diode structure (Fig. 1c) that facilitates the delivery of charge carriers to the defect for charge state switching. The charge state dynamics of a single NV center were investigated by time-resolved measurements and nondestructive single-shot readout of the charge state. Fast charge state switching rates (from negative to neutrally charged defects), which were greater than $0.72 \pm 0.10 \mu\text{s}^{-1}$ were realized. Furthermore, in no-operation mode, the realized charge states were stable for presumably much more than 0.45 s. We believe that the results obtained are useful not only for ultrafast electrical control of qubits, long T_2 quantum memory, and quantum sensors associated with single NV centers but also for classical memory devices based on single atomic storage bits working under ambient conditions.

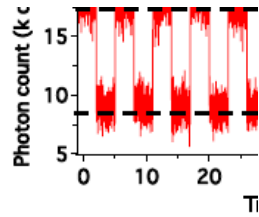


Fig. 2, Real-time trace of the fluorescence intensity with current switching between 0 and 0.2 mA. On the upper side, the 532-nm irradiation laser light and timing of the injected current pulse is shown. The fluorescence intensity histogram is shown in the right. The solid red line was obtained from fitting of the two Gaussian distributions. The blue straight line indicates the threshold of the two dynamical steady states and charge states.

(2) Pure negatively charged state of NV center in n-type diamond (submitted to Phys. Rev. X)

Negatively charged nitrogen-vacancy centers (NV^-) are promising for use as solid-state qubits and sensors in fields such as quantum information, magnetometry, and biosensing. Optical illumination is crucial for controlling and detecting NV^- qubits but inevitably causes stochastic charge-state transitions between NV^- and neutral charge states. Therefore, depending on the wavelength, such illumination decreases the steady-state NV^- population to 5%–80% of the total charge-state population in intrinsic diamond (Fig. 3a,b), which seriously degrades the optical and spin properties of the material. Here, we show that doping of phosphorus generates about $99.4 \pm 0.1\%$ NV^- under 1 μW and 593 nm excitation which is close to maximum absorption efficiency of NV^- (Fig. 3c,d). Under excitation at 593 nm, the luminescence of the pure NV^- increases five-fold, and the optically detected magnetic resonance signal increases three-fold compared with NV^- in intrinsic diamond. These results significantly enhance versatile potential of NV^- .

Generating a pure state including the charge state, close to 100 % NV^- population, is very important for quantum information applications. Studies involving the pre-selection and reset of the charge state were carried out to achieve high-fidelity operation in spite of the instability of the charge states. However, this approach makes scaling up of diamond quantum registers more challenging. Furthermore, single-shot readout of a nuclear spin indicate that the spin-flip probability of the conditional gate operation decreases because of the stochastic charge-state transitions. In addition, such charge-state transitions lead to spectral diffusion of the zero-phonon line of NV^- , which reduces the efficiency of two-photon quantum interference. For nanoscale sensing applications, it is crucial to keep NV^- stable; however, NV^- near the surface is unstable. In addition, high-contrast fluorescence switching between pure bright NV^- and the pure dark state (NV^0) is also very important for super-resolution microscopy.

Previously, NV charge states were controlled by heavy neutron irradiation, surface termination, and combined optical and electrical operations. From single-shot charge-state measurements, deterministic control from NV^- to NV^0 was realized by a purely electrical operation. Doping with nitrogen is one way to control the NV^- population. Nitrogen donors (P1 centers) can donate electrons to NV^0 , thereby changing its state to NV^- , because the activation energy ($E_A = 1.7$ eV) of P1 is less than the energy difference between the acceptor level, labeled ($-/0$), of the NV center

and the conduction-band edge. Recently, charge states of ensemble NV centers were modulated by ion-implantation of phosphorus and boron atoms. However, pure NV^- charge state has not yet been realized. Based on the activation energy, phosphorus doping during chemical vapor deposition (CVD) appears promising because that of phosphorus ($E_A = 0.57$ eV) is much less than that of P1.

In the present study, we quantitatively investigate the charge-state population of NV centers by using single-shot readout measurements in phosphorus-doped n-type diamond.

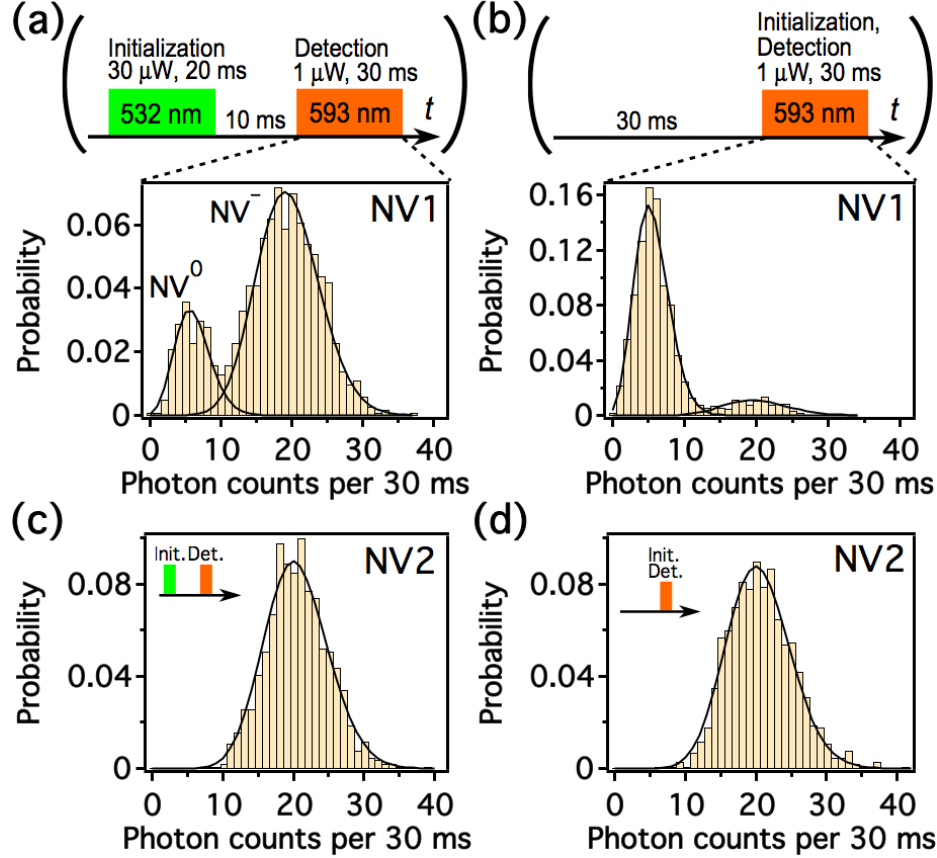


Fig. 3: Nondestructive single-shot charge-state measurements with two types of charge-state initialization: (a) 30 μW , 532 nm illumination and (b) 1 μW , 593 nm illumination. NV1 population exhibits a double Poisson distribution that is due to two different charge states. Initialization at 593 nm drastically decreases the population of the NV^- charge state. (c), (d) Conversely, NV2 has a single peak at the same position as NV^- irrespective of initialization conditions. We repeated each sequence 1000 times for each histogram.

(3) Electrical excitation of silicon-vacancy centers in single crystal diamond (submitted to Appl. Phys. Lett.)

Electrically driven emission from negatively charged silicon-vacancy (SiV^-) centers in single crystal diamond is demonstrated. The SiV centers were generated using ion implantation into an i region of a p-i-n single crystal diamond diode. Both electroluminescence and the photoluminescence signals exhibit the typical emission that is attributed to the $(\text{SiV})^-$ centers. Under forward and reversed biased PL measurements, no signal from the neutral $(\text{SiV})^0$ defect could be observed. The realization of electrically driven $(\text{SiV})^-$ emission is promising for scalable nanophotonics devices employing color centers in single crystal diamond.

Color centers in diamond are promising building blocks for a variety of applications, including quantum information processing and integrated nanophotonics. Recently, a major research focus has been dedicated to identify bright single photon emitters with narrow line widths and fast excited state lifetimes at the near infrared spectral range. One emerging candidate is the negatively charged silicon vacancy (SiV^-) color center that consists of an interstitial silicon splitting two vacancies in the diamond lattice. The SiV exhibits photostable room temperature operation with narrow zero phonon line (ZPL) (

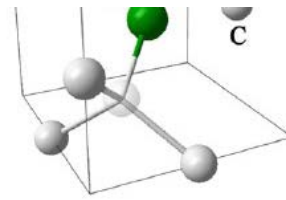


Fig. 4, Structure of SiV center in diamond.

$\sim 1\text{-}3\text{ nm}$), fast excited state lifetime ($\sim 1\text{ ns}$), polarized excitation and optical readout of its spin state at low temperature. These excellent photophysical properties are therefore attractive for applications spanning bio-imaging, nanophotonics and quantum information processing.

Although the SiV centers have been studied in details under optical excitation, electroluminescence properties of this center in a single crystal diamond remain unexplored. Electrical excitation is important since it opens pathways to engineer scalable devices employing electrically driven emitters and realize efficient packaging on a single chip. Furthermore, it enables studying charge injection and dynamics between various charge states of a particular defect. In this work we report on electrical excitation of engineered SiV defects in a single crystal diamond. In particular, we show that the same emission is obtained using optical and electrical excitation - a unique feature that has not been demonstrated in single crystalline diamond yet.

List of Publications and Significant Collaborations that resulted from your AOARD

supported project: In standard format showing authors, title, journal, issue, pages, and date, for each category list the following:

- a) papers published in peer-reviewed journals,
- b) papers published in peer-reviewed conference proceedings,
- c) papers published in non-peer-reviewed journals and conference proceedings,
- d) conference presentations without papers,
- e) manuscripts submitted but not yet published, and
- f) provide a list any interactions with industry or with Air Force Research Laboratory scientists or significant collaborations that resulted from this work.

Attachments: Publications a), b) and c) listed above if possible.

DD882: As a separate document, please complete and sign the inventions disclosure form.

Important Note: If the work has been adequately described in refereed publications, submit an abstract as described above and refer the reader to your above List of Publications for details. If a full report needs to be written, then submission of a final report that is very similar to a full length journal article will be sufficient in most cases. This document may be as long or as short as needed to give a fair account of the work performed during the period of performance. There will be variations depending on the scope of the work. As such, there is no length or formatting constraints for the final report. Keep in mind the amount of funding you received relative to the amount of effort you put into the report. For example, do not submit a \$300k report for \$50k worth of funding; likewise, do not submit a \$50k report for \$300k worth of funding. Include as many charts and figures as required to explain the work.

List of Publications and Significant Collaborations that resulted from your AOARD supported project: In standard format showing authors, title, journal, issue, pages, and date, for each category list the following:

a) papers published in peer-reviewed journals,

- [1] **Journal name:** Phys. Rev. X, 4, 01107 (2014).
Title: “Deterministic electrical charge state initialization of single nitrogen-vacancy center in diamond”
Date: 2014/4/1
Authors: Y. Doi, T. Makino, H. Kato, D. Takeuchi, M. Ogura, H. Okushi, H. Morishita, T. Tashima, S. Miwa, S. Yamasaki, J. Wrachtrup, Y. Suzuki, N. Mizuochi,
- [2] **Journal name:** Appl. Phys. Lett., 105, 261601 (2014).
Title: “Atomistic mechanism of perfect alignment of nitrogen-vacancy centers in diamond.”
Date: 2014/12/29
Authors: T. Miyazaki, R. Miyamoto, T. Makino, H. Kato, S. Yamasaki, T. Fukui, Y. Doi, N. Tokuda, M. Hatano, N. Mizuochi,
- [3] **Journal name:** Appl. Phys. Express. 7, 055201 (2014).
Title: “Perfect selective alignment of nitrogen-vacancy center in diamond”,
Date: 2014/4/9
Authors: T. Fukui, Y. Doi, T. Miyazaki, R. Miyamoto, H. Kato, T. Matsumoto, T. Makino, S. Yamasaki, R. Morimoto, N. Tokuda, M. Hatano, Y. Sakagawa, H. Morishita, T. Tashima, S. Miwa, Y. Suzuki, N. Mizuochi,
- [4] **Journal name:** Nature communications. 5, 3424 (2014).
Title: “Observation of dark states in a superconductor diamond quantum hybrid system”
Date: 2014/4/8
Authors: X. Zhu, S. Saito, Y. Matsuzaki, R. Amsüss, K. Kakuyanagi, T. Shimo-oka, N. Mizuochi, A. Saitoh, K. Nemoto, W. J. Munro, K. Semba,
- [5] **Journal name:** Nature Materials, 13, 50–56 (2014).
Title: “Highly sensitive nanoscale spin-torque diode”,
Date: 2014/1
Authors: S. Miwa, S. Ishibashi, H. Tomita, T. Nozaki, E. Tamura, K. Ando, N. Mizuochi, T. Saruya, H. Kubota, K. Yakushiji, T. Taniguchi, H. Imamura, A. Fukushima, S. Yuasa Y. Suzuki,
- [6] **Journal name:** Solid State Commun. 183, 18 (2014).
Title: “Spin-dependent tunneling in magnetic tunnel junctions with Fe nanoparticles embedded in MgO matrix”
Date: 2014/4/1
Authors: P. V. Thach, S. Miwa, Do Bang, T. Nozaki, F. Bonell, E. Tamura, N. Mizuochi, T. Shinjo and Y. Suzuki,
- [7] **Journal name:** Phys. Rev. Lett. 114, 120501 (2015).
Title: “Improving the coherence time of a quantum system via a coupling with an unstable system”,
Date accepted: 2015/1/25
Authors: Y. Matsuzaki, X. Zhu, K. Kakuyanagi, H. Toida, T. Shimo-oka, N. Mizuochi, K. Nemoto, K. Semba, W. J. Munro, H. Yamaguchi, S. Saito
- [8] **Journal name:** J. Appl. Phys. accepted.
Title: “Voltage modulation of propagating spin waves in Fe”,
Date accepted: 2014/11
Authors: K. Nawaoka, Y. Shiota, S. Miwa, H. Tomita, E. Tamura, N. Mizuochi, T.

b) papers published in peer-reviewed conference proceedings,

None

c) papers published in non-peer-reviewed journals and conference proceedings,

None

d) conference presentations without papers,

- [1] **Conf. name:** 28th Diamond Symposium, Tokyo, Japan
Title: "Perfect selective alignment of nitrogen-vacancy center in diamond"
Date: 2014/11/19
- [2] **Conf. name:** APES-IES-SEST2014),the Joint Conference of APES2014, IES and SEST2014, Nara, Japan
Title: "Electric Field Dependence of Coherence Time of the Electron Spin in the Diamond NV Center"
Date: 2014/11/14
- [3] **Conf. name:** The 75th Autumn Meeting, 2015 of The Japan Society of Applied Physics, Hokkaido, Japan
Title: "Perfect selective alignment of nitrogen-vacancy center in diamond"
Date: 2014/9/17
- [4] **Conf. name:** 25th International Conference on Diamond and Carbon Materials (DCM2014), Madrid, Spain
Title: "Perfect selective alignment of nitrogen-vacancy center in diamond"
Date: 2014/9/8
- [5] **Conf. name:** 25th International Conference on Diamond and Carbon Materials (DCM2014), Madrid, Spain
Title: "Deterministic charge state control of single nitrogen vacancy center in diamond"
Date: 2014/9/8
- [6] **Conf. name:** The 15th IUMRS-International Conference in Asia, Kyushu, Japan
Title: "Perfect alignment of the orientation of nitrogen-vacancy center in diamond"
Date: 2014/8/25
- [7] **Conf. name:** 14th Asian Quantum Information Science Conference, Kyoto, Japan
Title: "Deterministic charge state control of single NV center in diamond with p-i-n diamond diode "
Date: 2014/8/22
- [8] **Conf. name:** 2014 Autumn meeting of the Physical Society of Japan
Title: "Electric field dependence of electron spin states in diamond NV centers"
Date: 2014/9/9
- [9] **Conf. name:** 69th Annual meeting of the Physical Society of Japan
Title: "Electrical Charge State Control of Single NV center in Diamond "
Date: 2014/3/28
- [10] **Conf. name:** 69th Annual meeting of the Physical Society of Japan
Title: "Initialization of single nuclear spin of NV center in diamond"
Date: 2014/3/28
- [11] **Conf. name:** 69th Annual meeting of the Physical Society of Japan
Title: "The role of quantum memory in a distributed quantum computing"

Date: 2014/3/27

- [12] **Conf. name:** The 61st the Japan Society of Applied Physics Spring meeting
Title: "Optically detected magnetic resonance spectroscopy of diamond with a high density of nitrogen-vacancy centers"
Date: 2014/3/18
- [13] **Conf. name:** The 61st the Japan Society of Applied Physics Spring meeting
Title: "Electric Field Effect on NV Centers in Diamond under High Electric Field"
Date: 2014/3/18

e) manuscripts submitted but not yet published, and

- [1] **Journal name:** Phys. Rev. A, submitted.
Title: "Improving the lifetime of the NV center ensemble coupled with a superconducting flux qubit by applying magnetic fields"
Date received: 2014/11/4
Authors: Y. Matsuzaki, X. Zhu, K. Kakuyanagi, H. Toida, T. Shimo-oka, N. Mizuochi, K. Nemoto, K. Semba, W. J. Munro, H. Yamaguchi, S. Saito
- [2] **Journal name:** J. Appl. Phys., submitted
Title: "Inverse spin-Hall effect in NiFe|p-type diamond"
Date received: 2014/12/11
Authors: N. Fukui, H. Morishita, S. Kobayashi, S. Miwa, T. Shinjo, N. Mizuochi, Y. Suzuki
- [3] **Journal name:** Phys. Rev. X, submitted
Title: "Pure negatively charged state of NV center in n-type diamond"
Date received: 2014/12/8
Authors: Y. Doi, T. Fukui, H. Kato, T. Makino, S. Yamasaki, S. Miwa, F. Jelezko, Y. Suzuki, N. Mizuochi
- [4] **Journal name:** Nature Communications, submitted
Title: "A Germanium-Vacancy Single Photon Source in Diamond"
ArXiv: <http://arxiv.org/abs/1503.04938>
Date received: 2015/3/16
Authors: T. Iwasaki, F. Ishibashi, Y. Miyamoto, Y. Doi, S. Kobayashi, T. Miyazaki, K. Tahara, K. Jahnke, L. Rogers, B. Naydenov, F. Jelezko, S. Yamasaki, S. Nagamachi, N. Mizuochi, M. Hatano
- [5] **Journal name:** Appl. Phys. Lett., submitted
Title: "Electrical excitation of silicon-vacancy centers in single crystal diamond"
ArXiv: <http://arxiv.org/abs/1503.04778>
Date received: 2015/3/11
Authors: A. M. Berhane, S. Choi, H. Kato, T. Makino, N. Mizuochi, S. Yamasaki, I. Aharonovich
- [6] **Journal name:** AIP Advances, submitted
Title: "Growth of perpendicularly magnetized CoFeB thin films on a polymer buffer and voltage-induced change of magnetic anisotropy at the MgO|CoFeB interface"
Date received: 2015/2/25
Authors: D. D. Lam, F. Bonell, Y. Shiota, S. Miwa, T. Nozaki, E. Tamura, N. Mizuochi, T. Shinjo, Y. Suzuki, S. Yuasa

- [7] **Journal name:** New J Phys., submitted
Title: “Analysis of the spectroscopy of a hybrid system composed of a superconducting flux qubit and diamond NV- centers”
Date received: 2015/2/16
Authors: Y. Matsuzaki, H. Cai, K. Kakuyanagi, H. Toida, X., Zhu, N. Mizuochi, K. Nemoto, K. Semba, W. Munro, S. Saito, H. Yamaguchi
- [8] **Journal name:** Appl. Phys. Lett., submitted
Title: “Control of coherence among a single electron spin and three nearest neighbor ¹³C nuclear spins of a NV center in diamond”
Date received: 2015/1/26
Authors: T. Shimo-Oka, H. Kato, S. Yamasaki, S. Miwa, Y. Suzuki, N. Mizuochi
- [9] **Journal name:** Appl. Phys. Lett., submitted
Title: “Large voltage-induced magnetic anisotropy field change in ferrimagnetic FeGd”
Date received: 2015/1/13
Authors: K. Tanaka, S. Miwa, Y. Shiota, N. Mizuochi, T. Shinjo, Y. Suzuki

f) provide a list any interactions with industry or with Air Force Research Laboratory scientists or significant collaborations that resulted from this work.

4. Invited talks (event name, title, date):

- [1] **Event name:** 59th Annual Magnetism & Magnetic Materials Conference (MMM), Hawaii, USA
Title: “Single spin and charge manipulation of NV center in diamond”
Date: 2014/11/5
- [2] **Conf. name:** The 15th IUMRS-International Conference in Asia, Kyushu, Japan
Title: "Single photon, spin and charge manipulation of diamond register "
Date: 2014/8/25
- [3] **Event name:** The 75th Autumn Meeting, 2015 of the Japan Society of Applied Physics, Hokkaido, Japan, Symposium “Frontier of Spintronic Materials and Devices”
Title: Quantum spintronics by NV center in diamond
Date: 2014/9/18
- [4] **Event name:** Okinawa School in Physics: Coherent Quantum Dynamics, Okinawa, Japan
Title: "Quantum information and Metrology by NV center in diamond"
Date: 2014/9/16
- [5] **Event name:** The Magnetism Society of Japan, Tokyo, Japan
Title: "Magnetic sensor by using NV center in diamond"
Date: 2014/7/11
- [6] **Event name:** JSPS photo-electronics 132 committee, 292nd symposium, Tokyo, Japan
Title: "Single photon source by NV center in diamond"
Date: 2014/5/12
- [7] **Event name:** 12th symposium on photo-electronics for young researchers
Title: "Single photon source and spin manipulation by NV center in diamond"
Date: 2014/4/19

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5. Award for best paper, best poster (title, date):

- [1] **Title:** "Gold Young Scholar Award" in 25th International Conference on Diamond and Carbon Materials. "Deterministic charge state control of single nitrogen vacancy center in diamond"
Y. DOI (Doctor course student of Mizuochi' lab.)
(<http://www.materialstoday.com/materials-chemistry/news/award-winners-from-dcm-2014/>)
Date: 2014.9.10
- [2] **Title:** The paper of "Perfect selective alignment of nitrogen-vacancy center in diamond" in Applied Physics Express was selected as "SPOTLIGHTS" research. (<http://iopscience.iop.org/1882-0786/page/Spotlights>)
Date: 2014.4.10

6. Award of fund received related to your research efforts (name, amount, date):

- [1] **Name:** Strategic Information and Communications R&D Promotion Programme (Ministry of internal affairs and communications), "Research on diamond NV center for next generation quantum information device."
Amount: \$193,700 at 2014
Date: 2014.4.1
- [2] **Name:** CREST: Core Research for Evolutional Science of Japan Science and Technology Agency, "Revolutional magnetic sensor for bio-systems based on carbon nanomaterials"
Amount: \$158,167 at 2014
Date: 2014.4.1
- [3] **Name:** Consignment research of National institute of information and communications technology, "Research on technology of quantum repeater"
Amount: \$91,000 at 2014
Date: 2014.4.1
- [4] **Name:** Toray foundation, "Research on Quantum information device"
Amount: \$35,000 at 2014
Date: 2014.4.1
- [5] **Name:** Bilateral Joint Research Projects (Japan-Germany) (Japan society for the promotion of science), "Development of multi-qubit spintronics for quantum communication"
Amount: \$22,917 at 2014
Date: 2014.4.1